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INSTRUMENTS FOR ANALYSIS OF GASES ACCORDING TO THEIR MAGNETIC PROPERTIES

(A Survey)

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[Figures and table have not been reproduced but are available in the original in CIA.]

For the purpose of evaluating the magnetic properties of weakly-magnetic substances it is usual to employ the magnitude of magnetic susceptibility. A distinction is made between the volumetric magnetic susceptibility:

$$\varkappa = \frac{I}{H} \qquad (1)$$

(where I is the intensity of magnetization equal to the magnetic moment per unit volume of the body, and H is the intensity of the magnetic field) and the specific susceptibility:

$$\chi = \frac{\varkappa}{d} \qquad (2)$$

where d is the density of the substance.

For paramagnetic substances the values of the above-mentioned susceptibilities are positive; for diamagnetic substances they are negative.

Table 1 gives the values of volumetric magnetic susceptibility for gases at a temperature of 20 degrees Centigrade [1].

The specific magnetic susceptibility does not depend upon the pressure (or in the case of diamagnetic gases, upon temperature). In the case of paramagnetic gases the following dependence, determined from Curie's Law, generally holds:

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$$\chi = \frac{C}{T} \tag{3}$$

where C is Curie's constant and T is the absolute temperature.

The dependence of a gas' density d upon pressure P and temperature T is expressed by the equation:

$$PV_1 = RT = P\frac{M}{d}, \tag{3'}$$

where V_1 is the volume of one mole of the gas; R is the gas constant; M is the molecular weight of the gas. Hence the density of the substance is expressed thus:

$$d = \frac{PM}{RT}. \tag{3''}$$

Substituting the expression for density into Formula 2 and solving for volumetric magnetic susceptibility, we obtain the following formulas:

a. for diamagnetic gases:

$$\varkappa = \frac{\chi M}{R} \cdot \frac{P}{T}; \tag{4}$$

b. for paramagnetic gases: $\varkappa = \frac{CM}{R} \cdot \frac{P}{T^2}$. (5)

From Table 1 it is obvious that oxygen possesses the greatest magnetic susceptibility; after oxygen follow nitrous oxide and nitrogen dioxide.

The magnetic properties of the remaining gases are considerably lower.

The magnetic susceptibility of a mixture of gases equals the sum of the partial magnetic susceptibilities possessed by the components of the mixture; basically, it is determined by the contents of O_2, NO, or NO_2. From this it follows that the concentration of one of these three gases in a mixture can be determined from the magnetic properties of the mixture.

This method of gas analysis has found the widest application in industrial and laboratory practice. It is exceptionally convenient and important from the practical standpoint for the determination of oxygen in mixtures. The latter use is clarified by the following applications:

1. Oxygen is applied widely in diverse fields of industry [2, 3].

2. The known methods of chemical analysis of oxygen mixtures are not convenient for the purposes of continuous control and regulation of processes [4, 5].

3. The physical properties of oxygen (except its magnetic susceptibility) differ only slightly from the properties of most other gases with which oxygen is usually found in mixture; it is this fact that makes difficult the application of other accepted physical methods of analysis.

4. The high magnetic susceptibility of oxygen ensures, in the majority of cases, the certainty of the results of analysis and also ensures great sensitivity.

5. Analysis that is based upon the difference in magnetic properties of the components making up a mixture is convenient and adaptable to continuous automatic control and regulation of industrial processes.

The absolute values of magnetic susceptibility of gases are very small; therefore measurement must be based upon procedures that ensure the very highest sensitivity.

Most known procedures utilize one or another physical phenomenon that permits one to determine indirectly the magnetic susceptibility of the gas. To such phenomena belong the following:

1. A body located in a nonuniform magnetic field, when surrounded by a paramagnetic gas, undergoes an apparent decrease in magnetic susceptibility.

2. The viscosity of a paramagnetic gas decreases in a magnetic field.

3. The thermal conductivity of a paramagnetic gas decreases in a magnetic field.

4. Around a heated body surrounded by a paramagnetic gas, a thermomagnetic convection will take place in a nonuniform magnetic field thereby causing the body to cool.

Below we give a detailed analysis of each phenomenon and a description of the various types of instruments.

1. Instruments of the Mechanical Type

The force acting upon a body of volume V located in a nonuniform magnetic field is determined, as is well known, by the following expression:

$$F=\int_0^V (\varkappa - \varkappa_r)\, H\frac{dH}{dx}\, dV, \qquad (6)$$

where $\varkappa$ is the volumetric magnetic susceptibility of the body; $\varkappa_r$ is the volumetric magnetic susceptibility of the gas surrounding the body; H is the intensity of the magnetic field; the derivative dH/dx is the gradient of field strength H with respect to the direction of the force acting on the body.

In this case we employ a law quite analogous to Archimedes' Law: the magnetic susceptibility of the body suffers an apparent decrease by an amount equal to the susceptibility of the medium in which the body is placed [6].

Just as it is possible to determine the specific weight of a liquid by the loss in weight of a body immersed in a liquid, it is equally possible to determine the magnetic susceptibility of a gas surrounding a body by measuring mechanical forces acting upon the body.

An instrument for determination of oxygen in gaseous mixtures that is based upon the utilization of this principle, has been worked out by P. L. Kapitsa [7]. In this instrument a glass rotor in the form of a long body of ellipsoidal cross-section is suspended between the poles of a powerful permanent magnet (see Figure 1). The poles are given a shape that will ensure the maximum nonuniformity or irregularity of the field in the gap. The gas mixture to be analyzed passes through a tube surrounding the rotor. Depending upon the magnetic susceptibility of the gas mixture, the rotor turns through a large or small angle. The reading of the angle is by a light beam.

In order to damp the oscillations of the rotor a short-circuited coil of copper wire is fastened to the rotor; damping currents are generated in this coil during the swinging of the rotor. The instrument can be adjusted for zero readings. In order to do this the coil is disconnected and the ends of the winding are brought outside through the suspension filaments. From an external source a direct current is supplied to the coil, which establishes the zero position for the rotor. The magnitude of the current necessary for compensation is a measure of the magnetic susceptibility of the gas. By a suitable inclusion of resistance thermometers it is possible to compensate for temperature errors.

However, only measurement by the method of deflection finds practical application. In order to eliminate temperature errors the instrument is inclosed in a thermostat.

A method has been proposed for the compensation of temperature errors which permits to dispense with the thermostat. [8] In this case the ends of the coil winding are led outside, as for zero readings, and a differential thermocouple is connected to the coil. One soldered junction of the thermocouple measures the temperature of the gas; the other junction is kept at a constant temperature (melting ice). The thermo-electric currents, which are proportional to the temperature of the gas, generate mechanical forces which correct for the temperature error.

Another design is known for the instrument of the mechanical type. [9] A glass rotor in the form of two hollow spheres, fastened at the ends of a small rod, is suspended by a quartz filament (see Figure 2). The spheres are placed between two pairs of poles of a magnet (see Figure 3). This instrument uses two horseshoe-shaped magnets made of alnico-V alloy, weighing 125 grams each.

Instruments of the mechanical type are utilized to determine oxygen in gas mixtures. Their readings are proportional to the partial pressure of the oxygen. In order to determine the percentual concentration it is necessary to introduce a correction for the atmospheric pressure in Forumla (5). It is also possible to employ automatic compensation in measurements of pressure: for example, it is possible to employ the constructions described below in Section 4.

The instruments are prepared for various intervals of measurement of partial pressures from 0-20 to 0-800 mm/Hg. The accuracy of measurement is around one percent.

A disadvantage of instruments of this type (mechanical) is their sensitivity to vibrations, a fact which limits their field of application. At the present time these mechanical type instruments are being utilized mainly in laboratory practice. However, their use in industry under suitable conditions is not excluded.

In conjunction with photorelays these instruments can serve to regulate industrial processes.

2. Variation in Viscosity of Gases in a Magnetic Field

The influence of a magnetic field upon the viscosity of paramagnetic gases has been shown repeatedly by experimental investigations [10, 11]. The dependence found for oxygen is represented graphically by the curves in Figure 4. The viscosity of a gas decreases with increase in the magnetic field strength H up to a certain definite limit. At a constant temperature the decrease in viscosity depends upon the ratio H/P.

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Physically the phenomenon can be explained by the fact that the magnetic field changes the direction of the axes of rotation of the molecules. The probability of collision of the molecules and, consequently, the length of the free path depend upon the angle between the axis of rotation of the molecule and the direction of its motion. [12]

As seen from the curves of Figure 4, the variation in viscosity is not considerable in absolute magnitude. At atmospheric pressure and for a field strength H = 6000 oersteds this variation is only 0.2 percent. Because of this, the phenomenon is not being applied in the analysis of gases.

3. Instruments Based upon Variation of Thermal Conductivity of Gases in a Magnetic Field

The thermal conductivity of gases, according to the molecular-kinetic theory, is determined by the following formula:

$$\lambda = \varepsilon c_v \eta. \qquad (6')$$

where eta is the gas viscosity; epsilon is a coefficient, which depends upon the atomicity of the gas; C_v is the thermal capacity of the gas at constant volume.

Since the action of the magnetic field causes a decrease in the viscosity of a gas, it is necessary to expect a simultaneous decrease in the thermal conductivity also. This phenomenon was experimentally verified and has been investigated in detail. [13]

The variation in thermal conductivity, as a function of the magnetic field strength H and gas pressure P, is analogous to the variation in viscosity (Figure 5), exceeds it approximately 1.8 times on the basis of a comparison of absolute values.

The maximum variation in thermal conductivity is 1.18 percent. However, this quantity can be attained only for small pressures or for strong fields.

It has been found empirically that the variation in thermal conductivity satisfies the law following:

$$-\frac{\Delta\lambda}{\lambda} = \frac{aX^2}{1+bX+cX^2}, \qquad (7)$$

$$X = \frac{H}{P}\sqrt{T} \qquad (8)$$

where a, b, c are constant coefficients; H is the magnetic field strength; P is the pressure in millimeters of mercury; and T is the absolute temperature.

The molecular-kinetic theory of gases gives the following relations:

$$\sqrt{T} = c_1 v; \quad P = c_2 n v^2; \quad l = c_3/n; \quad t_s = \frac{l}{v}, \qquad (8')$$

where C_1, C_2, and C_3 are the coefficients constant for a given gas; v is the average speed of the molecules; n is the number of molecules per unit volume; l is the length of free path of the molecules; t_s is the time between two collisions of one molecule.

Substituting the relations introduced above into Equation (8), we obtain:

$$\frac{H}{P}\sqrt{T} = H\frac{c_1 v}{c_2 n v^2} = H\frac{c_1 c_3 l}{c_2 v} = \frac{c_1 c_3}{c_2} H t_s. \qquad (8'')$$

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The variation in thermal conductivity under the influence of a magnetic field depends upon the time between two collisions of a molecule. Obviously, the time necessary for the turning of the axis of rotation of a molecule under the influence of magnetic field is commensurate with the time t_s. For large pressures and small values of the magnetic field strength, the process of reorientation of molecules will not have sufficient time to be completed during the free flight of a molecule. This explains the nature of the dependences expressed in Figure 5.

Figure 6 shows the dependence of the variation in thermal conductivity upon the angle between the magnetic lines of force and the direction of heat flow.

Instruments have been constructed for the industrial analysis of gases; these instruments are based upon the above-described phenomenon. [1'] Figure 7 gives schematic representation of an instrument of this type, illustrating the principles involved. The difference in the resistances of heated filaments included in two arms of a bridge is measured. One of the filaments is placed between the poles of a permanent magnet giving a strong and uniform magnetic field.

According to data from the literature, this instrument permits one to measure the content of O_2 with an accuracy up to 0.01 percent. It is necessary to assume that such accuracy can be attained only under the most ideal conditions. The presence of even small quantities of hydrogen must lower considerably the sensitivity and accuracy of this instrument. In order to eliminate undesirable displacements of the filament due to the interaction of the current flowing in it with the magnetic flux, the bridge is operated by means of an alternating current of high frequency. Use is also made of filaments of the bifilar type which are sealed in quartz. This permits the employment of direct current. In order to eliminate errors that are caused by variations in the external temperature, the instrument is inclosed in a thermostat.

4. Instruments Based upon Utilization of Phenomenon of Thermomagnetic Convection

The phenomenon of thermomagnetic convection has been submitted to detailed experimental study. [15]

Convection appears around a heated body such as a platinum filament placed in a nonuniform magnetic field and surrounded by a paramagnetic gas. The particles of gas located around the filament are heated and thus partially lose their own magnetic properties, according to the law expressed by Formula (5). Then they are expelled from the magnetic field by the colder gas particles, just as in the instruments of the mechanical type the spheres of the rotor are expelled.

If one designates the temperature of the cold gas T_0 and the temperature of the heated gas around the filament by T, then from Formulas (5) and (6) we obtain the magnitude of the force with which the element of volume dV of heated gas is expelled;

$$dF = \frac{CMP}{R} H \frac{dH}{dx}\left(\frac{1}{T_0^2} - \frac{1}{T^2}\right) dV. \tag{9}$$

As a result of the action of these forces, a continuous convective motion of the gas takes place around the filament, causing a lowering of the filament's temperature.

The curves show in Figure 8 the variation in the resistance of a platinum wire (diameter 0.1 mm, length 7 mm, enclosed in a glass vessel with oxygen) for various displacements of the wire between the poles of the magnet. One of the poles of the magnet and the vessel are schematically drawn. The displacement was in the horizontal direction.

It is obvious from the curves that when a filament is located near the edges of a pole, where the product $H(dH/dx)$ is at a maximum, the resistance of the filament falls sharply. The amount of decrease in resistance reaches 4.3 percent (for $H = 20,000$ oersteds and $dH/dx = 20,000$ oersteds per centimeter).

In Figure 9, similar curves are drawn for displacements of the vessel in vertical direction. The influence of natural thermal convection can be seen from the curves. When the filament is located under the poles between the points A and B, the current of thermomagnetic convection is directed downward and is opposed to the current of thermal convection. At point B they balance each other and the filament possesses maximum excess heating.

For the same reason, the maximum reduction in resistance at point D is less than under the pole at point F, where both convective currents flow in the same direction.

It is obvious from Formula (9) that the effect of cooling of the filament depends not upon the absolute value of the magnetic susceptibility of the gas, but upon the temperature coefficient of susceptibility; that is, upon the derivative $d\varkappa/dT$.

Formula (8) is derived for paramagnetic gases, the magnetic properties of which satisfy Curie's Law (3); that is, for gases satisfying the law: $\varkappa \equiv 1/T^2$. For diamagnetic gases the following law holds: $\varkappa \equiv 1/T$. It is clear that thermomagnetic convection in these gases will be considerably weaker. NO and NO_2 also do not follow Curie's Law and, as shown by Richardson's experiment [16], the cooling effect on the filament is 12 percent for NO, and 3.3 percent for NO_2, of the same effect for O_2, while their magnetic susceptibilities are 45.2 percent for NO, and 6.2 percent for NO_2, of the susceptibility of O_2. This fact and also the considerate absolute magnitude of the cooling effect on the filament permits one to apply this effect successfully for industrial use in the determination of oxygen in gaseous mixtures. It is known that several instruments have been completed that are based upon this phenomenon.

Figure 10 shows the cross-sectional view of a transmitting element of one of these instruments; Figure 11 is its main circuit diagram. [17] Four platinum heating filaments connected into a bridge are located in the common chamber of the instrument. Two of these are placed near the pole of a magnet; the other two are placed outside the magnetic field.

The gas to be analyzed is blown through the chamber. The supply for the bridge is provided by a 20 kilocycle oscillator. The voltage from the diagonal of the bridge is amplified and is supplied to the measuring or recording apparatus.

In order to eliminate temperature errors the transmitting element of the instrument is enclosed in a thermostat having a temperature of 52 degrees centigrade.

The pressure of the gas is maintained at a constant level by means of the apparatus indicated schematically in Figure 11. The apparatus includes a differential manometer with a condenser transmitting element, which measures the pressure in front of and after the diaphragm, and an oscillator and frequency relay which controls the motor of the regulating valve.

The constructional data for this instrument are: Diameter of the platinum filaments, 0.01 mm; resistance, 15 ohms; voltage on bridge, about 3 volts; current through bridge, 0.18 ampere; temperature of filament, 260°C. The strength of the field in the gap is 8-10,000 oersteds. The rate of flow of the gas through the transmitting element is 100 cubic centimeters per minute.

Figure 12 shows the dependence of voltage in the diagonal of the bridge upon the oxygen content. Figure 13 shows the calibration curve for the instrument.

Figures 14 and 15 show the circuit and cross-sectional view of the transmitting element of an instrument of different design. [16] The characteristic feature of this instrument is the use of a heater in the form of a bifilar spiral winding sealed in glass. This construction permits the use of alternating current for the bridge supply.

This instrument has two identical heaters placed in two different chambers. One of them is located in the magnetic field; the other is used for the purpose of compensation. The bridge has a device for automatic balancing which consists of an amplifier for the unbalance currents and a motor connected with the slide arm of a rheochord and a recording instrument.

Linearity of the calibration curve for the instrument is ensured by a suitably profiled cam.

As is obvious from Figure 15, gas exchange in the chamber proceeds exclusively by diffusion. This causes a certain lag in the indicator readings (in 15 seconds after a change in the gas composition the indicator goes 90 percent of the way between the old value and the new value). Compensation is provided in the instrument for errors caused by variations in humidity of the gas, temperature, and pressure. A porous plug which is continuously wetted is placed in the lower part of each chamber. The gas in passing close to this wetplug becomes saturated with water vapor. The temperature errors are eliminated by the use of a thermostat (at 55°C). In order to eliminate errors due to pressure variations, a device is provided as shown in Figure 14. A corrugated box, called a 'silfon', which is filled with air and hermetically sealed, is heated by an electric heater. The latter is controlled by contacts kinematically connected with the 'silfon'. The length of the 'silfon', depending upon its temperature and the external pressure, always remains constant; consequently the temperature of the 'silfon' is a function of the pressure. The thermometer, incorporated in a suitable manner in the scheme of the instrument, introduces temperature corrections.

The design data of the instrument is as follows: The heater is a platinum wire 0.025 mm in diameter; its resistance is 65 ohms. The magnetic field strength is 13,000 oersteds. The sensitivity of the instrument is about 3 millivolts on the diagonal of the bridge per 1 percent oxygen (in the first half of the scale).

Figure 16 gives a curve showing the variation in resistance of the heater in ohms as a function of the oxygen content. The instrument can be tested for its "zero" reading when the gas contains no oxygen. To do this, a permanent magnet is set on a hinge and can be swung out of the chamber.

Other types of instruments, which utilize the phenomenon of thermomagnetic convection, are also well known.

Of interest is the instrument illustrated schematically in Figure 17. The gas to be analyzed is divided into two symmetrical currents and passes by a transverse horizontally-situated tube which supports two heated windings, or coils, 1 and 2. These windings are connected to the measuring bridge supplied with direct current. Around the edge of winding 2 are located the poles of magnet 3. By virtue of the phenomenon described above, when a paramagnetic gas is present, a current of gas appears in the horizontal tube and this current is directed to the right from the magnetic pole. In consequence of the motion of the gas, winding 1 is cooled and winding 2 is heated, which fact results in an unbalancing of the bridge. Such a constructional feature has a number of advantages over those described previously: (1) the readings of the instrument are less influenced by the thermal conductivity of the gaseous mixture; (2) the instrument does not need alternating current for the supply; (3) the instrument is simpler and more dependable from the viewpoint of construction and exploitation.

The deficiency of this instrument is the large mass of the body to be heated, which leads to a considerable lag in the indicator readings. An instument of this type, just as those described above, must be used with a thermostat and must be compensated for variations in barometric pressure.

Soviet instruments of this type are manufactured for use in the control of blast furnace gases in metallurgical plants, see M. M. Faynberg's article [18].

This instrument gives an output of about 120 millivolts for a variation of oxygen content from 0 to 100 percent and for a resistance of each heating element amounting to 25 ohms. The lag in indicator reading is about 90 seconds.

5. Fields of Application of Instruments

Instruments for the analysis of gas, usually oxygen, that are based upon the measurement of magnetic properties have just been introduced. Operational results have not yet been accumulated to a sufficient extent to permit a proper evaluation of the possibility of a wide-scale use of these instruments. It can be stated, however, that the instruments of the mechanical type will be employed mainly in the laboratory in view of their sensitivity to vibration. In certain cases under favorable conditions they can also be used in industry as control or regulating devices.

The instruments that are based upon the measurement of the thermal conductivity of gases in a magnetic field, in comparison with instruments based upon the utilization of the thermomagnetic convection effect, possess obvious advantages only in two cases: (1) in the analysis of rarefield gases under a pressure of less than 300 millimeters of mercury, and (2) in the analysis of mixtures containing NO or NO_2 (in the absence of O_2).

In the analysis of oxygen under a pressure higher than 300 mm/Hg, the instrument of the second type (nonmechanical) is more convenient, since it gives a greater relative variation in resistance.

Both types of instruments can be used in the laboratory and in industry.

It is possible to mention the following definite areas of application where the instruments based upon the utilization of thermomagnetic convection can be used as control devices or transmitting elements in automatic systems:

1. Chemical plants [5] .

2. Air separation installations;

3. At boiler plants and metallurgical furnaces, in the analysis of stack gases and regulation of combustion [4]

4. On blast furnaces, for control and regulation of the oxygen-air mixture in enriched blasting [2, 3]

5. Analysis of the composition of air in mine shafts, tunnels, submarines, pressure chambers (barokamera), sewers, wells, etc.

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